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(71) Applicant:
Nisshinbo Industries, Inc.
Chuo-ku, Tokyo 103-8650 (JP)

(72) Inventors:
• Saito, Kazuo,
c/o Nisshinbo Ind. Inc.
Adachi-ku, Tokyo 123-0843 (JP)
• Hagiwara, Atsushi,
c/o Nisshinbo Ind. Inc.
Adachi-ku, Tokyo 123-0843 (JP)

(74) Representative:
Laufhütte, Dieter, Dr.-Ing. et al
Lorenz-Seidler-Gosse
Widenmayerstrasse 23
80538 München (DE)

(54) Material for fuel cell separator and fuel cell separator made from said material

(57) The present invention provides:

a material for fuel cell separator obtained by molding a mixture containing at least a conductive powder and a binder which is a rubber-modified phenolic resin, and
a fuel cell separator made from the above material.

The material for fuel cell separator alleviates the problems of the prior art, is superior in impact resistance or tenacity, causes no rupture or the like when made into a thin plate and used in a fuel cell, and is equivalent to conventional products in density and electrical properties.

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Description**BACKGROUND OF THE INVENTION**5 **(1) Field of the Invention**

[0001] The present invention relates to a material for fuel cell separator and a fuel cell separator made from said material. More particularly, the present invention relates to a material for fuel cell separator, superior in impact resistance or tenacity, as well as to a fuel cell separator made from said material.

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10 **(2) Description of the Prior Art**

[0002] Fuel cells are advantageous in that they require substantially no fossil fuel (whose depletion in the near future is feared) and, when used for power generation, generate substantially no noise and are high in energy recovery as compared with other methods for power generation. Therefore, fuel cells are being developed as a power generation plant of relatively small size for buildings and factories.

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[0003] Among fuel cells, a solid polymer type fuel cell operates at lower temperatures than other types of fuel cell do; therefore, it has such characteristics that not only the parts constituting the cell are little corroded, but also the cell can discharge a relatively large electric current for the low temperature operation. Therefore, the solid polymer type fuel cell is drawing attention as a substitute electric source for internal combustion engine of vehicle.

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[0004] Among the parts constituting the above solid polymer type fuel cell, the separator is generally a flat plate having a plurality of parallel grooves (function as flow channels) formed at one or both sides, and has a role of transferring the electricity generated at the gas diffusion electrode of fuel cell, to the exterior and also a role of discharging the water formed in the grooves in the course of electricity generation, to reliably use each groove as a path for a reactant gas which is allowed to flow into the fuel cell.

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[0005] As fuel cells have become lighter and thinner in recent years, it has become necessary that the separator therefor be made thinner as well. However, conventional fuel cell separators or materials therefor have been unable to satisfy this necessity.

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[0006] That is, conventional fuel cell separators are obtained, for example, by machining a graphite plate impregnated with a thermosetting resin, or by molding an expanded graphite sheet into a separator shape; these fuel separators or materials therefor have low impact resistance or tenacity when made as a thin plate and, as a result, break when used in a fuel cell or, even if no such breakage occurs, are unusable per se in fuel cells for automobile or in portable fuel cells (wherein use conditions may be sever).

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SUMMARY OF THE INVENTION

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[0007] The present invention aims at providing a material for fuel cell separator, which alleviates the above-mentioned problems of the prior art, is superior in impact resistance or tenacity, causes no breakage or the like when made into a thin plate and used in a fuel cell, and is equivalent to conventional products in density and electrical properties; and a fuel cell separator made from the material.

[0008] According to the present invention, there are provided:

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a material for fuel cell separator obtained by molding a mixture containing at least a conductive powder and a binder which is a rubber-modified phenolic resin; and
a fuel cell separator made from the above material.

DETAILED DESCRIPTION OF THE INVENTION

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[0009] As the conductive powder used in the present invention, there can be mentioned, for example, a powder of a carbon material typified by natural graphite (e.g. scaly graphite or lumpy graphite), artificial graphite, acetylene black, carbon black, Ketjen black, expanded graphite or the like. There is no particular restriction as to the kind of the conductive powder as long as the powder is conductive.

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[0010] The conductive powder can have an average particle diameter of, for example, 10 nm to 100 µm, preferably, 3 µm to 80 µm. When the average particle diameter is 10 nm or more, excellent formability is achieved, and when the average particle diameter is 100 µm or less, excellent conductivity is achieved.. The conductive powder may be subjected, as necessary, to a hydrophilization treatment by control of firing conditions or by using a chemical, a gas or the like. These conductive powders can be used singly or in admixture of two or more kinds.

[0011] The conductive powder is molded together with a binder to become a material for fuel cell separator, of the

present invention. In the present invention, a rubber-modified phenolic resin is used as the binder.

[0012] The rubber-modified phenolic resin can be obtained by reacting an unvulcanized rubber with a phenolic resin. As the unvulcanized rubber, there can be mentioned fluororubber, silicone rubber, butyl rubber, chloroprene rubber, nitrile rubber, nitrile-chloroprene rubber, chlorinated butyl rubber, chlorinated polyethylene, epichlorohydrin rubber, epichlorohydrin-ethylene oxide rubber, epichlorohydrin-ethylene oxide-acryl glycidyl ether terpolymer, urethane rubber, acrylic rubber, ethylene-propylene rubber, styrene rubber, butadiene rubber, natural rubber, etc. These rubbers can be used singly or in admixture of two or more kinds.

[0013] The rubber-modified phenolic resin can have a rubber modification degree of 10 to 90%, preferably, 10 to 30%. When the rubber modification degree is 10% or more, the rubber-modified phenolic resin is low in elastic modulus and a separator improved in impact resistance can be obtained. When the rubber modification degree is 90% or less, particularly 40% or less, the properties of the resin used for modification are not changed largely and the resulting separator is not low in strength.

[0014] Incidentally, the rubber modification degree can be determined by the following formula:

$$15 \quad [(weight\ of\ unvulcanized\ rubber)/(weight\ of\ unvulcanized\ rubber + weight\ of\ phenolic\ resin)] \times 100$$

[0015] The material for fuel cell separator, of the present invention can be obtained by molding the conductive powder and the rubber-modified phenolic resin as binder. The proportions of the conductive powder and the rubber-modified phenolic resin can be 100 parts by weight (the conductive powder) and 5 to 50 parts by weight (the rubber-modified phenolic resin). When the proportion of the rubber-modified phenolic resin is 5 parts by weight or more, the resulting material for fuel cell separator can have high impact resistance. When the proportion of the rubber-modified phenolic resin is 50 parts by weight or less, the resulting material can give a fuel cell separator reliably having a required conductivity.

[0016] In molding the conductive powder and the rubber-modified phenolic resin to produce a material for fuel cell separator, of the present invention, these two raw materials are mixed first. The mixing can be conducted by a known industrial method such as stirring rod, kneader, ball mill, sample mill, mixer, static mixer, ribbon mixer or the like. In this case, in order to expect improved molding, the rubber-modified phenolic resin may be dissolved in an appropriate solvent, followed by granulation.

[0017] The thus-obtained mixture of the conductive powder and the rubber-modified phenolic resin is shaped into a desired molding, i.e. a material for fuel cell separator, of the present invention. This shaping can be conducted by a known molding method such as pressure molding, hydrostatic pressure molding, extrusion molding, injection molding, belt press, press molding, press heating, roll pressing or the like, or by a combination of the above two or more molding methods.

[0018] The temperature employed in shaping the mixture is determined depending upon the resin used in the mixture, but can be, for example, ordinary temperature to 400°C. The pressure employed in shaping can be, for example, 100 to 250 kg/cm². In order to chemically stabilize the molding obtained, the molding may be heat-treated.

[0019] The fuel cell separator of the present invention is produced from the above-obtained material for fuel cell separator, of the present invention. In general, the fuel cell separator has paths (function as a flow field) for gas flow, grooves for discharging the water generated in the electricity-generating reaction of cell, etc.; therefore, the fuel cell separator can be produced by forming such paths, grooves, etc. in the above-mentioned shaping step for the material for fuel cell separator, or by forming, after the shaping step, the paths, grooves, etc. using an appropriate means such as machining or the like.

[0020] The thus-obtained fuel cell separator of the present invention has an elastic modulus in bending, of 40 to 1 GPa and a deflection at rupture in bending test, of 0.1 to 3 mm. Therefore, the fuel cell separator causes no rupture or the like when made in a thin plate and used in a fuel cell.

[0021] Incidentally, the elastic modulus was measured according to JIS K 6911. That is, a test piece of 100 mm in length, 4±0.2 mm in height and 10±0.5 mm in width was produced; the test piece was supported by two fulcrums apart by a distance of 64±0.5 mm; a load (a pressure wedge) was applied to the test piece at the center of the two fulcrums; loads and deflections (mm) were measured up to the time when the test piece was broken; and a load-deflection curve was prepared. The elastic modulus of the test piece was calculated from the following formula.

$$E_f = (L_v^3 / 4Wh^3) \times (F/Y) \times (1/1000)$$

wherein

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 E_f = elastic modulus in bending (GPa),
 L_v = length (distance) between fulcrums (mm),
 W = width of test piece (mm).

h = height of test piece (mm), and

F/Y = gradient of the straight-line portion of load-deflection curve (N/mm)

[0022] Moreover, the fuel cell separator of the present invention has a bulk density of 1.7 to 2.0 g/cm³, a resistivity of 5 to 40 mΩ · cm and a Shore hardness of 2 to 30; thus, the present fuel cell separator is equivalent to conventional products in density and electrical properties.

[0023] The present invention is described in more detail below by way of Examples.

Examples 1 to 5

[0024] A scaly graphite powder (as conductive powder) having an average particle diameter of 30 µm and an acrylic rubber-modified phenolic resin having a rubber modification degree shown in Table 1 were mixed in proportions shown in Table 1, for 10 minutes using a mixer. The resulting mixture was placed in a mold of 200 mm x 200 mm and molded into a separator shape under the conditions of mold temperature = 160°C, molding pressure = 150 kg/cm² and molding time = 5 minutes. The molding obtained was measured for bulk density, resistivity, elastic modulus in bending, deflection at rupture in bending test, and Shore hardness. Separately, a fuel cell separator having a thickness of 1.0 mm was produced in the same manner and used in a fuel cell; and the condition of the separator in the cell was observed. The results are shown in Table 1.

Examples 6 and 7

[0025] A fuel cell separator was produced in the same manner as in Example 1 or 5 except that the acrylic rubber-modified phenolic resin was changed to a silicone rubber-modified phenolic resin having a rubber modification degree of 20%. The separator was measured for properties in the same manner as in Examples 1 to 5, and also the condition of the separator when used in a fuel cell was observed. The results are shown in Table 1.

Table 1

Examples	1	2	5	1	5	6	7
Conductive carbon powder	100	100	100	100	100	100	100
Acrylic rubber-modified phenolic resin	30	30	30	30	10		
Silicone rubber-modified phenolic resin						30	10
Rubber modification degree	10	20	30	90	20	20	20
Bulk density(g/cm ³)	1.85	1.78	1.77	1.70	1.78	1.81	1.82
Resistivity(mΩ · cm)	30	30	29	29	12	19	9
Elastic modulus in bending(GPa)	25	8	4	1	7	18	15
Deflection at rupture in bending test (mm)	0.15	0.40	0.80	1.00	0.20	0.20	0.15
Shore hardness	29	25	12	10	10	11	13
Condition when used in fuel cell	Good						

Comparative Example 1

[0026] A fuel cell separator was produced in the same manner as in Example 1 that the acrylic rubber-modified phenolic resin was changed to a phenolic resin. The separator was measured for properties in the same manner as in Examples 1 to 5, and also the condition of the separator when used in a fuel cell was observed. The results are shown in Table 2.

Comparative Example 2

[0027] A fuel cell separator was produced in the same manner as in Example 1 that no acrylic rubber-modified phenolic resin was used. The separator was measured for properties in the same manner as in Examples 1 to 5, and also the condition of the separator when used in a fuel cell was observed. The results are shown in Table 2.

Table 2

Comparative Examples	1	2
Conductive carbon powder	100	100
Phenolic resin	30	
Bulk density (g/cm ³)	1.95	1.90
Resistivity(mΩ · cm)	35	5
Elastic modulus in bending(GPa)	45	50
Deflection at rupture in bending test (mm)	0.08	0.04
Shore hardness	40	100
Condition when used in fuel cell	Assembling was impossible due to appearance of cracks during assembling	Same as left

[0028] As described above, the fuel cell separator of the present invention is produced from the material for fuel cell separator, of the present invention obtained by molding a mixture containing at least a conductive powder and a binder which is a rubber-modified phenolic resin; is superior in impact resistance or tenacity, causes no rupture or the like when made as a thin plate and used in a fuel cell, and is equivalent to conventional products in density and electrical properties.

Claims

1. A material for fuel cell separator obtained by molding a mixture containing at least a conductive powder and a binder which is a rubber-modified phenolic resin.
2. A material for fuel cell separator according to Claim 1, wherein the proportion of the rubber-modified phenolic resin is 5 to 50 parts by weight per 100 parts by weight of the conductive powder.
3. A material for fuel cell separator according to Claims 1 or 2, wherein the rubber modification degree of the rubber-modified phenolic resin is 10 to 90%.
4. A material for fuel cell separator according to Claim 1, wherein the average particle diameter of the conductive powder is 10 nm to 100 µm.
5. A fuel cell separator made from a material set forth in any of Claims 1 to 3.
6. A fuel cell separator according to Claim 5, having an elastic modulus in bending, of 40 to 1 GPa and a deflection at rupture in bending test, of 0.1 to 3 mm.
7. A fuel cell which uses the separator according to Claims 5 or 6.



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(71) Applicant: Nissinbo Industries, Inc.
Chuo-ku, Tokyo 103-8650 (JP)

(72) Inventors:

- Saito, Kazuo, c/o Nissinbo Ind. Inc.
Adachi-ku, Tokyo 123-0843 (JP)
- Hagiwara, Atsushi, c/o Nissinbo Ind. Inc.
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Lorenz-Seidler-Gossel
Widenmayerstrasse 23
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EUROPEAN SEARCH REPORT

Application Number

EP 99 11 3726

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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim							
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<p>The present search report has been drawn up for all claims</p> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 33%;">Place of search</td> <td style="width: 33%;">Date of completion of the search</td> <td style="width: 34%;">Examiner</td> </tr> <tr> <td>THE HAGUE</td> <td>4 December 2003</td> <td>GONZALEZ-CUENCA, M</td> </tr> </table>				Place of search	Date of completion of the search	Examiner	THE HAGUE	4 December 2003	GONZALEZ-CUENCA, M
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<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>									

**ANNEX TO THE EUROPEAN SEARCH REPORT
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